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Noisy Time-Dependent Spectra

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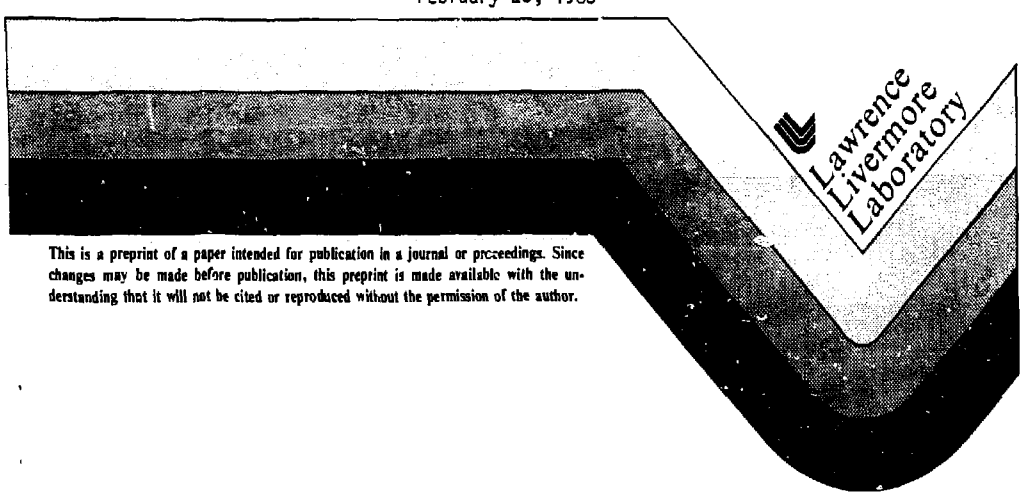

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Abstract

The definition of a time-dependent spectrum registered by an idealized spectrometer responding to a time-varying electromagnetic field as proposed by Eberly and Wodkiewicz and subsequently applied to the spectrum of laser-induced flourescence by Eberly, Kunasz, and Wodkiewicz is here extended to allow a stochastically fluctuating (interruption model) environment: we provide an algorithm for numerical determination of the time-dependent fluorescence spectrum of an atom subject to excitation by an intense noisy laser and interruptive relaxation.

1. Introduction

Observations of light scattered by atoms and molecules provide important clues to underlying atomic or molecular structure. With an intense, nearly monochromatic, light source tuned near a resonance frequency the resulting fluorescence radiation reveals significant properties of the light-matter interaction--most notably AC Stark splitting of the fluorescence spectral line[1]. The opportunity provided by tunable pulsed lasers to examine transient excitation effects prompted recent reexamination of the theory of time-dependent spectra[2], with particular application to laser-induced fluorescence[3]. The wealth of possible transient phenomena motivates study of time-dependent spectra; the dramatic alteration of spectra from strongly excited atoms[4] motivates systematic treatment of the resonant atom-radiation system.

Although isolated atoms interacting with monochromatic light provide an instructive theoretical model, real atoms encounter a fluctuating

environment of perturbing interactions and real lasers undergo a variety of fluctations in phase, frequency, and amplitude. Incorporation of such phenomena into dynamical equations[5] by way of empirical relaxation times and bandwidths provides a significant step toward realistic modeling, but it is desirable to base theory upon more fundamental ground when possible. A vast literature exists treating stochastic processes and their effects upon the contours or spectra lines observed in emission or absorption. Few papers have discussed stochastic effects in fluorescence, and applications to time-dependent fluorescence remain unexplored. The present paper addresses this topic.

2. Time-Dependent Spectra

Discussions of the notion of spectra provide numerous alternative resolutions of the basic time-frequency uncertainty needed for a definition of time-dependent spectra[2]. We adopt the definition of Eberly and Wodkiewicz[2]: the (normalized) physical spectrum registered at time t by an idealized spectrometer of bandwidth γ centered about the spectrometer frequency ω_s of an electric field whose positive-frequency part is the complex-valued function $\varepsilon(t)$ is

$$I(t) = N\cdot 2\gamma \int_{t_0}^{t} dt' \exp\left[-(\gamma - i\omega_s)(t-t')\right] \times \int_{t_0}^{t} dt'' \exp\left[-(\gamma + i\omega_s)(t-t'')\right] \varepsilon(t')\,\varepsilon^*(t'') \tag{2.1}$$

for a field first recorded at time t_0. Here N is a normalizing constant incorporating detector characteristics and units of measurement.

Our interest lies with electromagnetic fields radiated from atomic (or molecular) dipoles. Following well-known procedures[6] we can relate the electric field entering the spectrometer to time variations of the atomic dipole-moment source. For a two-state atom this procedure, taken with the symmetry property

$$\varepsilon(t')\varepsilon^*(t'') = [\varepsilon(t'')\varepsilon^*(t')]^*$$

leads from (Eq. 2.1) to the integral

$$I(t) = 2\mathrm{Re}\ N' \cdot 2\gamma \int_{t_0}^{t} dt' \exp[-(\gamma - i\omega_s)(t-t')] \times \int_{t_0}^{t'} dt'' \exp[-(\gamma+i\omega_s)(t-t'')] \langle \sigma'_{21}(t')q'_{12}(t'')\rangle \tag{2.2}$$

where $\langle \ldots \rangle$ denotes an expectation value and $\sigma'_{ij}(t)$ is the time-developed Heisenberg operator[6] which at $t = 0$ is the "projector":

$$\sigma'_{ij} \equiv |i\rangle\langle j| \quad . \tag{2.3}$$

(The prime on σ' anticipates transformation (2.4) to a more convenient basis.) The normalization factor N' incorporates the magnitude of the atomic transition moment as well as source-detector geometry, thereby retaining the normalization of I(t).

Anticipating application to fluorescence of an atom excited by a laser having instantaneous carrier frequency ω_L (possibly variable) we introduce the rotating wave transformation

$$\sigma_{21}(t) = e^{-i\varphi(t)}\,\sigma'_{21}(t) \qquad \sigma_{22}(t) = \sigma'_{22}(t)$$
$$\sigma_{12}(t) = e^{+i\varphi(t)}\,\sigma'_{12}(t) \qquad \sigma_{11}(t) = \sigma'_{11}(t) \tag{2.4}$$

of the elementary Heisenberg operators (2.3). Here the instantaneous phase is

$$\varphi(t) \equiv \int_0^t dt'\, \omega_L(t').$$

To simplify expressions we define

$$D \equiv \omega_S - \bar{\omega}_L$$

to be the frequency offset between the spectrometer and a (constant) mean laser carrier frequency ω_L, and we define the laser phase-fluctuation function

$$\Phi(t'-t'') \equiv \int_{t''}^{t'} dt\, [\omega_L(t) - \bar{\omega}_L] \tag{2.5}$$

$$= \varphi(t') - \varphi(t'') - (t'-t'')\,\bar{\omega}_L \quad .$$

(This function vanishes if the laser frequency ω_L remains constant; we assume fluctuations are stationary in time so that Φ depends only on the interval $t'-t''$.) Then, with $\tau \equiv t'-t''$ and $N' = 1$, we write eqn. (2.2) as

$$I(t) = 2\mathrm{Re}\; 2\gamma \int_0^t d\tau\, e^{-(\gamma - iD)\tau} \int_{t_0}^{t-\tau} dt''\, e^{-2\gamma(t-t'')}$$
$$\times\, e^{i\Phi(\tau)} \langle \sigma_{21}(t''+\tau)\, \sigma_{12}(t'') \rangle \tag{2.6}$$

Formulas (2.2) and (2.6) for I(t) apply to idealized observation of a single atom. In practice we detect radiation from many atoms, each immersed in a slightly different environment of fluctuating perturbations. Studies of the dependence typically average many observations, each recorded the same time t after pulse initiation. Thus we require spectra $\bar{I}(t)$ averaged over a variety of randomly varying conditions.

The most direct approach to this goal considers an ensemble of excitation histories, each subject to a definite but randomized variation of conditions. If we let $\{X(t)\}$ denote the ensemble average of a function X(t), then the desired spectrum $\bar{I}(t)$ is obtained by computing individual functions I(t) and averaging these over all possible histories:

$$\bar{I}(t) = \{I(t)\}.$$

Such a straightforward approach facilitates Monte-Carlo computations in which one constructs cumulative averages of a succession of functions, each of which rests upon some definite history of randomized variation.

The present paper follows an alternative route in which we obtain, through the method of marginal averages, procedures for direct evaluation of the time dependent of stochastic averages such as $\{\langle\sigma_{21}(t'')\sigma_{12}(t')\rangle\}$. The spectrum I(t) then obtains from the integral of this averaged correlation function.

To maintain generality we consider the function

$$X \equiv X(t_0+\tau,t_0) = e^{i\Phi(\tau)}\langle x(\hat{t}_0+\tau)y(\hat{t}_0)\rangle \qquad (2.7)$$

where $\hat{x}(t)$ and $\hat{y}(t)$ are some arbitrary combination of atomic operators σ_{ij} (t)

and where

$$\dot{\phi} = \omega_L - \bar{\omega}_L$$

is the deviation of the instantaneous laser frequency ω_L from the constant mean value $\bar{\omega}_L$. With appropriate choice of x and y the desired spectrum I(t) is the real part of the integral

$$\bar{G}(t) = 4\gamma \int_0^t dt\, e^{-(\gamma - iD)\tau} \int_{t_0}^{t-\tau} dt''\, e^{-2\gamma(t-t'')}$$

$$x\{X(t''+\tau, t'')\} \qquad (2.8)$$

(Note that D does not fluctuate: the factor $e^{i\phi}$ in Eq. 2.7 compensates our use of a fluctuating frequency ω_L in the transformation 2.4 underlying the operators comprising x and y.)

In the steady-state limit the correlation function becomes independent of t"

$$\{X(t''+\tau, t'')\} \to \{X(\tau, 0)\}$$

and the integral over t" can then be evaluated; the subsequent limiting case of an infinitely sharp spectrometer ($\gamma \to 0$) observed over an infinite duration ($t_0 \to -\infty$, $t \to \infty$) then yields the conventional result

$$\bar{G} = 2\mathrm{Re} \int_0^\infty d\tau\, e^{-(\gamma - iD)\tau} \{X(\tau, 0)\} \qquad (2.9)$$

That is, the steady-state spectrum is the Fourier transform of the dipole autocorrelation function. [For a detailed discussion of the conflicting limits used here: steady-state, $\gamma \to 0$, infinite observation time, etc., see reference 6.]

Rather than limit consideration to steady-state conditions, we retain the more complicated formula (2.8); such a definition is necessary to examine transient atomic response following sudden illumination.

Evaluation of spectra requires a determination of the functional dependence of an atomic correlation function $\{X(t',t'')\}$ upon time, through evaluation of the time dependence of atomic operators $\hat{x}(t)$ and $\hat{y}(t)$ followed by subsequent stochastic averaging. Note that laser fluctuations affect X in two ways: explicitly through the phase $\Phi(t)$ of Eq. 2.4; and implicitly through stochastic properties of $\hat{x}(t)$ and $\hat{y}(t)$ induced by fluctuating fields.

In weak-field far-off-resonant steady-state excitation a linear relationsnip links the operator $\sigma_{12}(t)$ and the instantaneous field value $\varepsilon(t)$. Under such conditions the replacement

$$\{X(t',t'')\} \rightarrow N''\{\varepsilon^*(t')\varepsilon(t'')\} \tag{2.10}$$

reduces the problem to the determination of the electric-field correlation function. That is, in the absence of redistributive effects, the weak scattered light spectrum is the same as the incident spectrum.

However, the interesting regime of strongly excited fluorescence, when perturbation theory fails to account adequately for excitation, requires more detailed examination of the relationship between excitation field and dipole moment.

3. Atomic Time Dependence

The basic atomic excitation dynamics of a multilevel atom is governed by suitable generalization of the two-state operator Bloch equations in the Rotating Wave Approximation, which we write as follows

$$\frac{d}{dt}\sigma_{12} = -[\frac{A}{2}+i\Delta]\sigma_{12} - \frac{i}{2}\Omega[\sigma_{22} - \sigma_{11}]$$

$$\frac{d}{dt}\sigma_{21} = -(\frac{d}{dt}\sigma_{12})^* \qquad (3.1)$$

$$\frac{d}{dt}\sigma_{22} = -A\sigma_{22} - i\Omega^*\sigma_{12} + i\Omega\sigma_{21}$$

$$\frac{d}{dt}\sigma_{11} = -(\frac{d}{dt}\sigma_{22})$$

where A is the spontaneous emission rate,

$$\Delta \equiv \Delta(t) = \omega^\circ - \omega_L \qquad (3.2)$$

is the (possibly fluctuating) instantaneous detuning of the laser frequency ω_L from the atomic Bohr-transition frequency ω°, and

$$\Omega \equiv \Omega(t) = -\langle 2|\hat{\underset{\sim}{d}}\cdot\underset{\sim}{\varepsilon}|1\rangle/\hbar \qquad (3.3)$$

is the (instantaneous) Rabi frequency appropriate to atomic dipole moment $\hat{\underset{\sim}{d}}$ interacting with the electric field

$$\underset{\sim}{E}(t) = \mathrm{Re}[\underset{\sim}{\varepsilon}(t)\exp(i\omega_L t)]. \qquad (3.4)$$

Incidentally we note that the adiabatic ($\dot{\sigma}_{12} = 0$) weak-field ($\sigma_{11} = 1$, $\sigma_{22} = 0$) limit of these equations yields the proportionality

$$\sigma_{12}(t) = \frac{i\Omega(t)}{[A+2i\Delta]} \qquad (3.5)$$

needed for replacement (2.10). This equation holds when $T \gg 1$, where T is the relaxation time.

To simplify further analysis we regard the operator $\hat{x}(t)$ of Eq. 2.7 not as a single operator $\sigma_{ij}(t)$ but as an ordered set of such operators. One such choice of orderings, for a two-state atom, is the four-component vector whose transpose is

$$x^T \equiv (\hat{x}_1, \hat{x}_2, \hat{x}_3, \hat{x}_4) = (\sigma_{11}, \sigma_{21}, \sigma_{12}, \sigma_{22})$$

although alternative combinations such as $\sigma_{22} \pm \sigma_{11}$ and/or $\sigma_{12} \pm i\sigma_{21}$ often prove advantageous for computations. It now follows from the existence of a linear equation for the $\sigma_{ij}(t)$ that the operator components of $\hat{x}(t)$ satisfy a linear equation having the form

$$\boxed{\frac{d}{dt}\hat{x}_j(t) = -i \sum_k W_{jk}(t)\,\hat{x}_k(t)} \tag{3.11a}$$

or, more simply

$$\frac{d}{dt}\hat{x}(t) = -iW(t)\hat{x}(t). \tag{3.11b}$$

From these equations it follows that the $X(t,t_0)$ of definition (7) satisfies the equation

$$\frac{d}{dt}X(t,t_0) = -i[W(t) - 1\cdot\Phi(t)]\,X(t,t_0) \tag{3.12}$$

where 1 denotes the unit matrix.

The coefficient matrix $W(t)$ appearing in Eqs. (3.11) has elements proportional to the instantaneous Rabi frequency $\Omega(t)$ and the instantaneous laser-atom detuning $\Delta(t)$. Perturbations of the atomic energy levels caused by collisions appear here as time variations of the Bohr frequency ω° and, in turn, as fluctuations in $\Delta(t)$. Collisions altering atomic orientation relative to E or, alternatively, changes in laser polarization, produce changes in the phase of the complex-valued $\Omega(t)$. Fluctuations of laser amplitude or collision-induced atomic-state mixing produce variations in the magnitude of $\Omega(t)$. Changes in laser frequency can either be expressed as a time varying

phase of $\Omega(t)$ or, more commonly, in variations of both $\Phi(t)$ and $\Delta(t)$. We assume that such variations occur as fluctuations in a largely uncontrolled but statistically determined way. Thus $W(t)$ is a matrix whose elements are examples of stochastic processes, and Eqs. (3.12) are examples of multiplicative stochastic processes.[7]

A literature survey will reveal two classes of investigated Markovian stochastic processes in the theory of atomic excitation by lasers. The first category, that of Gaussian random processes[8] (an example is the phase diffusion model), requires only two-time correlation functions of the laser field, such as $\{\Omega(t)\Omega(t')^*\}$. This approach, though permitting exact solution in an interesting variety of cases, ultimately rest upon the assumption that underlying the interaction there is some variable that is delta correlated, i.e. is represented by white noise.

We shall here consider the second, less well-known, category that of jump processes developed in detail by Burshtein.[9,10] The jump or interruption model assumes that we can characterize the excitation dynamics by a set of parameter values α (e.g., values for magnitude and phase of the Rabi frequency Ω, the instantaneous detuning Δ and the laser fluctuation Φ). These remain constant except at discrete times $t_0 < t_1 < t_2 < \ldots$ when one or more parameters may take new values.

4. The Jump Model

We assume that the random processes are stationary. Thus we can define, independent of time, a probability $F(\alpha)$ of observing parameter values α. We assume that the processes are Markovian: values after a

jump depend, at most, only upon values immediately prior to the jump but not upon earlier history. Therefore, we can define a time-dependent conditional probability (transition probability) $f(\alpha,\beta)$ of observing parameter values α after a jump, given that the parameters had values β prior to the jump. These probabilities satisfy the equations

$$\int_{\alpha} f(\alpha,\beta) = 1 \tag{4.1}$$

$$\int_{\beta} f(\alpha,\beta)\, F(\beta) = F(\alpha). \tag{4.2}$$

Here the Schiff symbol $\int$ denotes summation over discrete variables and integration over continuous variables.

We assume that interruptions occur after random time intervals: as in radioactive decay the probability of remaining uninterrupted declines exponentially with time; we let T be the mean time between interruptions.

Note that, despite the introduction of a single unique mean interruption rate 1/T, it is possible to specify different characteristic time scales for phase and amplitude fluctuations; this we accomplish by our choice of the transition-probability matrix $f(\alpha,\beta)$.

5. Marginal Averages

Given these model-defining assumptions, we next examine the procedure needed to construct $\{X(t,t_0)\}$ as an average over all possible histories of parameter variation. The simplest such history involves no interruptions at all: parameter values remain fixed at the initial value α_0 subsequent to time t_0. This history produces the function

$$X^0(t|\alpha_0 t_0).$$

Similarly we define

$$X^1(t|\alpha_1 t_1, \alpha_0 t_0)$$

to be $X(t,t_0)$ when a single interruption occurs at time $t_1 > t_0$, with parameters α_0 changing to α_1 at this instant. More generally, we have an n-jump sequence

$$X^n(t|\alpha_n t_n,\ldots,\alpha_0 t_0)$$

in which parameters undergo changes $\alpha_0 \to \alpha_1 \to \ldots \alpha_n$ at times $t_0 < t_1 < \ldots t_n$. The desired average $\{X(t,t_0)\}$ is obtained by summing all such functions, for all possible parameter values and all possible interruption times, weighted by the appropriate probabilities.

The average over jump parameters consists of the summation

$$\sum_{\alpha_n}\sum_{\alpha_{n-1}} \cdots \sum_{\alpha_1}\sum_{\alpha_0} f(\alpha_n,\alpha_{n-1}) \cdots f(\alpha_1,\alpha_0)F(\alpha_0)$$

$$\times X^n(t|\alpha_n t_n, \ldots, \alpha_1 t_1, \alpha_0 t_0) \ .$$

The interruption-time average proceeds independently of parameter choices and requires the multiple integration

$$\int_{t_0}^{t}\frac{dt_n}{T}\int_{t_0}^{t_n}\frac{dt_{n-1}}{T}\cdots\int_{t_0}^{t_2}\frac{dt_1}{T}$$

$$\times \exp[-(\frac{t-t_n}{T})]\exp[-(\frac{t_n-t_{n-1}}{T})]\cdots\exp[-(\frac{t_1-t_0}{T})]$$

$$\times X^n(t|\alpha_n t_n,\alpha_{n-1}t_{n-1},\ldots,\alpha_1 t_1,\alpha_0 t_0).$$

The key elements in the Burshtein approach are the marginal averages $\{\ldots\}_\alpha^n$ and $\{\ldots\}_\alpha$ defined by the requirement that the full stochastic average $\{\ldots\}$ is the sum

$$\{X(t,t_0)\} = \sum_n \sum_\alpha \{X(t,t_0)\}_\alpha^n \qquad (5.1)$$

$$= \sum_\alpha \{X(t,t_0)\}_\alpha .$$

From the foregoing definitions we see that the marginal average $\{\ldots\}_\alpha^n$ is expressible as

$$\{X(t,t_0)\}_\alpha^n = \exp[-(\frac{t-t_0}{T})] \int_{t_0}^{t} \frac{dt_n}{T} \cdots \int_{t_0}^{t_2} \frac{dt_1}{T}$$

$$\times \sum_{\alpha_{n-1}} \cdots \sum_{\alpha_0} f(\alpha,\alpha_{n-1}) \cdots f(\alpha_1,\alpha_0)\, F(\alpha_0) \qquad (5.2)$$

$$\times X^n(t|\alpha\, t_n, \ldots, \alpha_0 t_0).$$

Fortunately, it is not necessary to actually evaluate this expression. Instead we can, as Zoller and Ehlotzky pointed out, use the property

$$X_n(t|\alpha_n t,\alpha_{n-1}t_{n-1},\ldots) = X^{n-1}(t|\alpha_{n-1}t_{n-1},\ldots) \qquad (5.3)$$

valid when $t = t_n$, together with the equation

$$\frac{d}{dt} X^n(t|\alpha t_n,\ldots) = -i[W(\alpha)-1\cdot\Phi(\alpha)]X^n(t|\alpha t_n,\ldots) \qquad (5.4)$$

where $W(\alpha)$ and $\Phi(t)$ are $W(t)$ and $\Phi(t)$ evaluated with the parameters α, to show from the definition (5.2) that

$$\frac{d}{dt} \{X(t,t_0)\}_\alpha^n = -\frac{1}{T}\{X(t,t_0)\}_\alpha^n \qquad (5.5)$$

$$- i[W(\alpha)-1\cdot\Phi(\alpha)]\ \{X(t,t_0)\}_\alpha^n + \frac{1}{T}\sum_\beta f(\alpha,\beta)\ \{X(t,t_0)\}_\beta^{n-1}.$$

It then follows that the marginal average $\{\ldots\}_\alpha$ satisfies the equation

$$\boxed{\frac{d}{dt}\{X(t,t_0)\}_\alpha = -i[W(\alpha)-\dot{\Phi}(\alpha)]\{X(t,t_0)\}_\alpha + \frac{1}{T}\sum_\beta [f(\alpha,\beta)-\delta\alpha\beta]\{X(t,t_0)\}_\beta .} \tag{5.6}$$

Given solutions to this equation, we carry out a final summation over α to obtain the integrand for Eqs. (2.8) or (2.9).

Note that when we write $W(t) = W(\alpha(t))$ needed for Eqn. (5.4) we make a strong assumption: we require that the Hamiltonian depend only upon the instantaneous values of all parameters and not upon the past history of any fluctuating parameter. This assumption is essential to the Bursntein method.

6. Solving the Bursntein Equation

Bursntein[9] examined several special cases of Eq. (5.6) which admit exact solution. We can, in principle, readily obtain a solution if we restrict consideration to discrete-valued parameters. Then the equation takes the form of a system of linear equations with constant coefficients. Recalling that $X(t,t_0)$ has components

$$X_j(t,t_0) \equiv e^{i\Phi(t-t_0)} \langle \hat{x}_j(t)\hat{y}(t_0)\rangle \quad , \tag{6.1}$$

we can write the equation as

$$\frac{d}{dt}\{X_j(t,t_0)\}_\alpha = -i \sum_{k\beta} M_{jk}(\alpha,\beta)\{X_k(t,t_0)\}_\beta \tag{6.2}$$

where

$$\boxed{M_{jk}(\alpha,\beta) = \delta_{\alpha\beta}\, W_{jk}(\alpha) + \delta_{jk}[-\delta_{\alpha\beta}\dot{\Phi}(\alpha) + \tfrac{i}{\hbar}(f(\alpha,\beta)-\delta_{\alpha\beta})]} \tag{6.3}$$

It then follows that a solution for $t > t_0$ can be written as

$$\{X_j(t,t_0)\}_\alpha = \sum_{k\beta} U_{jk}(\alpha,\beta|t-t_0)\ \{X_k(t_0,t_0)\}_\beta \tag{6.4}$$

where U satisfies the equation

$$\boxed{\frac{d}{dt}\, U_{jk}(\alpha,\beta|t) = -i \sum_{\ell\gamma} M_{k\ell}(\alpha,\gamma)\, U_{\ell k}(\gamma,\beta|t)} \tag{6.5}$$

subject to the initial condition

$$U_{jk}(\alpha,\beta|0) = \delta_{jk}\delta_{\alpha\beta}. \tag{6.6}$$

Now the objects of interest, here symbolized by components of $X(t,t_0)$, involve bilinear operator products such as $\sigma_{12}(t')\sigma_{21}(t'')$. Such bilinear forms satisfy the equal-time properties

$$\sigma_{ij}(t)\sigma_{k\ell}(t) = \delta_{ik}\sigma_{i\ell}(t) \quad . \tag{6.7}$$

We can apply these rules to construct a time-dependent matrix K such that

$$\boxed{\hat{x}_j(t)\hat{y}(t) = \sum_k K_{jk}\hat{x}_k(t)} \tag{6.8}$$

and in turn express the initial value of $X(t_0,t_0)$ as

$$X_j(t_0,t_0) = \langle \hat{x}_j(t_0)\hat{y}(t_0)\rangle \tag{6.9}$$

$$= \sum_k K_{jk} \langle x_k(t_0)\rangle \equiv \sum_k K_{jk}\gamma_k(t_0)$$

thereby defining a vector $Y(t)$. Upon taking marginal averages of this equation we obtain the result

$$\{X(t_o,t_o)\}_\alpha = K\{Y(t_o)\}_\alpha \quad . \tag{6.10}$$

Now $\{Y(t)\}_\alpha$ satisfies the same equation as $\{X(t,t_o)\}_\alpha$ except that $\Phi \equiv 0$. Hence it has solutions expressible in terms of an evolution matrix $U'(t)$. Thus we finally obtain the formulas

$$\{X_j(t_o+\tau,t_o)\} = \sum_{\substack{k\ell\beta\gamma \\ m\ \alpha}} U_{jk}(\alpha,\beta|\tau)K_{k\ell}U'_{\ell m}(\beta,\gamma|t_o)\{Y_m(0)\}_\gamma \tag{6.11a}$$

$$\{X_j(\tau,0)\} = \sum U_{jk}(\alpha,\beta|\tau)K_{km}\{\overline{Y}_m\}_\beta \tag{6.11b}$$

for the integrands of Eqns. (2.8) and (2.9) respectively.

The final term $\{Y_m(0)\}_\gamma$ of formula (6.11a) incorporates the initial atomic excitation conditions as well as the probability distribution for parameters:

$$\{Y_m(0)\}_\alpha = F(\alpha)\ \langle \hat{x}_m(0) \rangle \tag{6.12a}$$

For formula (6.11b), with its assumption of a steady state at time $t = 0$, we require the initial condition

$$\{\overline{Y}_m\}_\alpha = \lim_{t\to\infty} \sum_{j\beta} U_{mj}(\alpha,\beta|\tau)F(\beta)\ \langle \hat{x}_j(0) \rangle \tag{6.12b}$$

7. Eigenvector Methods

Given formulas (6.11) and the defining differential equations (6.5) for U it is, at least in principle, a straightforward matter to obtain explicit functional forms for the time dependence of U and thence

evaluate the integral for G(t). In the limit $t\to\infty$ these integrals become one-sided Laplace transforms, and solution of Eqs. (6.5) and (6.6) by Laplace transforms then provides immediately the desired spectral components. The presence of finite-time limits in the G(t) integrals makes transform techniques cumbersome and lends preference to the eigenvalue method. For this purpose it is useful to define some indexing scheme which maps the pair of discrete indices i, α (with i the component of atomic operator $\hat{x}_i$ and α the laser-interaction parameters) into a single discrete index, say $\underline{a}$. One has the reorganized indexing

$$U_{ij}(\alpha,\beta|t) \to U_{ab}(t)$$
$$M_{ij}(\alpha,\beta) \to M_{ab}$$

and Eqs. (6.5) and (6.6) read simply

$$\frac{d}{dt}\,U(t) = -iMU(t), \qquad U(0) = 1 \quad . \tag{7.1}$$

Following standard practice we can construct the required solutions U(t) from eigenvalues and eigenvectors (left and right) of the matrix M; given the eigenvalue equations

$$\boxed{\begin{array}{l}\sum_a [M_{ab} - \lambda\delta_{ab}] \langle b|\lambda\rangle = 0 \\ \\ \sum_a \langle\lambda|a\rangle[M_{ab} - \lambda\delta_{ab}] = 0\end{array}} \tag{7.2}$$

we write

$$U_{ab}(t) = \sum_{\lambda} \langle a|\lambda\rangle\, e^{-i\lambda t}\, \langle\lambda|b\rangle \tag{7.3}$$

or, in more detail,

$$U_{ij}(\alpha,\beta|t) = \sum_{\lambda} \langle i\alpha|\lambda\rangle\, e^{-i\lambda t}\, \langle\lambda|j\beta\rangle \quad . \tag{7.4}$$

Note that λ depends implicitly upon α: each choice of parameters α produces a new matrix M. Employing this result we express the average derived from Eqs. (6.11) as

$$\boxed{\{X_j(t_o+\tau,t_o)\} = \sum_{\lambda\lambda'} T_j(\lambda,\lambda')\, e^{-i\lambda\tau}\, e^{-i\lambda' t_o}} \tag{7.5a}$$

and

$$\boxed{\{X_j(\tau,0)\} = \sum_{\lambda} \bar{T}_j(\lambda)\, e^{-i\lambda\tau}} \tag{7.5b}$$

where

$$T_j(\lambda,\lambda') = \sum_{\alpha} \langle j\alpha|\lambda\rangle \sum_{k\ell\beta} \langle\lambda|k\beta\rangle\, K_{k\ell}\, \langle\ell\beta|\lambda'\rangle \tag{7.6a}$$

$$\times \sum_{m\gamma} \langle\lambda'|\gamma m\rangle\, \{Y_m(0)\}_{\gamma}$$

and

$$\bar{T}_j(\lambda) = \sum_{\alpha} \langle j\alpha|\lambda\rangle \sum_{k\ell\beta} \langle\lambda|k\beta\rangle\, K_{k\ell}\, \{\bar{Y}_{\ell}\}_{\beta} \quad . \tag{7.6b}$$

Upon substituting these expressions into the definitions (2.8-2.9) we obtain the final formulas

$$\bar{G}(t) = \mathrm{Re} \sum_{\lambda\lambda'} T(\lambda,\lambda')\, F_{\lambda\lambda'}(t) \tag{7.7a}$$

$$\bar{G} = \mathrm{Re} \sum_{\lambda} \bar{T}(\lambda)\bar{F}_{\lambda} \tag{7.7b}$$

where

$$F_{\lambda\lambda'}(t) = \frac{4q}{(\lambda-D+i\gamma)} \left[\frac{e^{-i\lambda' t} - e^{-i(\lambda-D-i\gamma)t}}{(\lambda-\lambda'-D-i\gamma)} + \frac{e^{-i\lambda' t} - e^{-2\gamma t}}{(\lambda' + 2i\gamma)} \right] \tag{7.8a}$$

and

$$\bar{F}_{\lambda} = \lim_{t\to\infty} \frac{1}{(\lambda-D+i\gamma)} \left[e^{-i\lambda t} - 1 \right] \tag{7.8b}$$

8. Summary of Method

We summarize the steps needed to obtain the time-dependent spectrum.

First we define our $\hat{x}(t)$ and $\hat{y}(t)$ operators as particular combinations of atomic operators $\sigma_{ij}(t)$. For example, we might choose

$$\hat{x} = (\hat{x}_1,\hat{x}_2,\hat{x}_3,\hat{x}_4)^T = (\sigma_{11},\sigma_{21},\sigma_{12},\sigma_{22})^T \tag{8.1}$$

$$\hat{y} = \sigma_{21}$$

so that the spectrum is the j=2 component of the $\bar{G}(t)$ integral:

$$\bar{I}(t) = \mathrm{Re}\, \bar{G}_2(t) \tag{8.2}$$

Using this definition and the equal-time product property of Eq. (6.7) we determine the matrix K: the choice (8.1) yields the result

$$\begin{bmatrix} \sigma_{11}\ \sigma_{12} \\ \sigma_{21}\ \sigma_{12} \\ \sigma_{12}\ \sigma_{12} \\ \sigma_{22}\ \sigma_{12} \end{bmatrix} = \begin{bmatrix} \sigma_{12} \\ \sigma_{22} \\ 0 \\ 0 \end{bmatrix} = \begin{bmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} \sigma_{11} \\ \sigma_{21} \\ \sigma_{12} \\ \sigma_{22} \end{bmatrix} \equiv K\,\hat{x} \quad . \tag{8.3}$$

From suitable generalization of the Bloch Eq. (3.1) we determine the functional dependence of W upon W and L; for definition (8.1) we have

$$\frac{d}{dt}\begin{bmatrix} \sigma_{11} \\ \sigma_{21} \\ \sigma_{12} \\ \sigma_{22} \end{bmatrix} = -\frac{i}{2}\begin{bmatrix} -iA & \Omega & -\Omega^* & 2i\gamma \\ \Omega^* & -W-iA & 0 & -\Omega^* \\ -\Omega & 0 & W-iA & \Omega \\ 0 & -\Omega & \Omega^* & -2iA \end{bmatrix}\begin{bmatrix} \sigma_{11} \\ \sigma_{21} \\ \sigma_{12} \\ \sigma_{22} \end{bmatrix} \tag{8.4}$$

$$\equiv -i\hat{W}\hat{x} \quad .$$

We next define the jump process: we choose the fluctuating parameters and assign values α, probability distributions $F(\alpha)$ and transition probabilities $f(\alpha,\beta)$; we select the mean time between interruption T.

Next we construct the array $M_{jk}(\alpha,\beta)$ defined in Eq. (6.3). For the present example this matrix has the elements

$$\begin{aligned} M_{11}(\alpha,\beta) &= -\dot{\Phi}(\alpha)\delta_{\alpha\beta} - \frac{i}{T}[f(\alpha,\beta) - \delta_{\alpha\beta}] \\ M_{12}(\alpha,\beta) &= [\tfrac{1}{2}\Omega(\alpha)]\delta_{\alpha\beta} \\ M_{13}(\alpha,\beta) &= [-\tfrac{1}{2}\Omega^*(\alpha)]\delta_{\alpha\beta} \\ &\vdots \\ M_{44}(\alpha,\beta) &= [-\dot{\Phi}(\alpha) - i\gamma]\delta_{\alpha\beta} + \frac{i}{T}[f(\alpha,\beta) - \delta_{\alpha\beta}] \end{aligned} \tag{8.5}$$

to be evaluated for an allowed values of α and β. Introducing

appropriate indexing we obtain eigenvalues λ and eigenvectors $\langle i\alpha|\lambda\rangle$ and $\langle\lambda|i\alpha\rangle$ of the matrix M.

Next we use these results to construct the vector $T(\lambda,\lambda')$ whose elements $T_j(\lambda,\lambda')$ are given by Eq. (7.6). For this purpose we require the initial-condition vector of Eq. (6.12). If the atoms are all initially unexcited, so that only the expectation value $\langle\sigma_{11}(0)\rangle = 1$ is nonzero, the formulas become

$$\{Y_m(0)\}_\alpha = \delta_{m,1}F(\alpha) \tag{8.6a}$$

$$\{\overline{Y}_m\}_\alpha = \sum_{\beta\lambda} \delta(\lambda,0)\ \langle m\alpha|\lambda\rangle\langle\lambda|\beta\rangle\ F(\beta) \tag{8.6b}$$

Given the functions $T_2(\lambda,\lambda')$ we evaluate $G_2(t)$ using formula (7.7a), and in turn we obtain I(t) from Eq. (8.2). Similarly we evaluate I from G_2 and $T_2(\lambda)$.

The foregoing procedure allows arbitrary fluctuations in laser phase, amplitude, and frequency as well as in atomic perturber environment. However, practical considerations of contemporary computer storage size limit the number of parameter values. A two state atom requires 4 indices j, so that if we allow 10 values for laser phase and 5 for laser amplitude the Matrix M has $(4\times10\times5)^2 = (200)^2$ entries. However, two-valued parameter-jump processes require much more modest storage.

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